

WHAT IS CLAIMED IS:

*Paul
A1*

1. A process for producing β -form tris-(2,3-epoxypropyl)-isocyanurate crystals containing from 2 to 15 wt% of α -form tris-(2,3-epoxypropyl)-isocyanurate in the interior of the crystals, which comprises the following steps (A), (B), (C) and (D):

(A) a step of reacting cyanuric acid with epichlorohydrin to form an addition product of cyanuric acid and epichlorohydrin, followed by dehydrochlorination to obtain a reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate,

(B) a step of removing epichlorohydrin from the reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate obtained in step (A), and dissolving the obtained tris-(2,3-epoxypropyl)-isocyanurate in a solvent,

(C) a step of gradually cooling the liquid obtained in step (B) at a cooling rate within 20 °C/hr for crystallization, followed by filtration to obtain crystals, and

(D) a step of washing and drying the crystals obtained in step (C).

2. The process according to Claim 1, wherein step (A) is a step of reacting (a) 1 mol of cyanuric acid, (b) from 5 to 180 mols of epichlorohydrin and (c) from 0.001 to 0.1 mol of at least one compound selected from the group consisting of a tertiary amine, a quaternary ammonium salt, a quaternary ammonium base, a tri-

substituted phosphine and a quaternary phosphonium salt,
as a catalyst, to obtain a reaction solution, adding from
2 to 6 mols of an alkali metal hydroxide or an alkali
metal alcoholate to the reaction solution for

5 dehydrochlorination, and then removing the resulting
alkali metal salt to obtain a reaction solution
containing tris-(2,3-epoxypropyl)-isocyanurate.

3. The process according to Claim 1, wherein the
solvent in which tris-(2,3-epoxypropyl)-isocyanurate is
10 dissolved in step (B) is acetonitrile, toluene, dioxane
or dimethylformamide.

4. The process according to Claim 1, wherein ultrasonic
waves are applied to the liquid in the process of
gradually cooling the liquid in step (C).

15 5. The process according to Claim 1, wherein the
washing in step (D) is carried out by using a solvent
capable of providing a solubility of at least 0.5 g/100 g
at 20°C to α -form tris-(2,3-epoxypropyl)-isocyanurate
and a solubility of less than 0.5 g/100 g at 20°C to β -
20 form tris-(2,3-epoxypropyl)-isocyanurate, in an amount of
from 0.5 to 10 times by weight relative to the β -form
tris-(2,3-epoxypropyl)-isocyanurate crystals.

6. The process according to Claim 1, wherein the
average particle size of the crystals obtained in step
25 (C) is from 20 to 500 μ m, and the drying in step (D) is
carried out under atmospheric pressure or under reduced
pressure in a gas stream at a temperature of from 120 to

G1
cont

00073766-0404

140°C.

a' cont
7. The process according to Claim 1, wherein the average particle size of the crystals obtained in step (C) is from 10 to 20 μm , and the drying in step (D) is
5 carried out under atmospheric pressure or under reduced pressure in a gas stream at a temperature of from 40 to 120°C.

8. A process for producing β -form tris-(2,3-epoxypropyl)-isocyanurate crystals containing from 2 to
10 15 wt% of α -form tris-(2,3-epoxypropyl)-isocyanurate in the interior of the crystals, which comprises the following steps (A), (B), (C') and (D):

(A) a step of reacting cyanuric acid with epichlorohydrin to form an addition product of cyanuric
15 acid and epichlorohydrin, followed by dehydrochlorination to obtain a reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate,

(B) a step of removing epichlorohydrin from the reaction solution containing tris-(2,3-epoxypropyl)-
20 isocyanurate obtained in step (A), and dissolving the obtained tris-(2,3-epoxypropyl)-isocyanurate in a solvent,

(C') a step of adding seed crystals to the liquid obtained in step (B) at a temperature lower by from 5 to 20°C than the temperature at which the liquid forms a
25 saturated solution, and gradually cooling the liquid at a cooling rate within 20 °C/hr for crystallization, followed by filtration to obtain crystals, and

(D) a step of washing and drying the crystals obtained in step (C').

a'
cont

9. The process according to Claim 8, wherein step (A) is a step of reacting (a) 1 mol of cyanuric acid, (b) from 5 to 180 mols of epichlorohydrin and (c) from 0.001 to 0.1 mol of at least one compound selected from the group consisting of a tertiary amine, a quaternary ammonium salt, a quaternary ammonium base, a tri-substituted phosphine and a quaternary phosphonium salt, as a catalyst, to obtain a reaction solution, adding from 2 to 6 mols of an alkali metal hydroxide or an alkali metal alcoholate to the reaction solution for dehydrochlorination, and then removing the resulting alkali metal salt to obtain a reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate.

10. The process according to Claim 8, wherein the solvent in which tris-(2,3-epoxypropyl)-isocyanurate is dissolved in step (B) is acetonitrile, toluene, dioxane or dimethylformamide.

11. The process according to Claim 8, wherein the addition of seed crystals in step (C') satisfies the following formulae (1) and (2):

$$1 \times 10^{10} \geq T \geq 1 \times 10^2 \quad (1)$$

$$T = 1.4 \times 10^{12} (m / (M \times D^3)) \quad (2)$$

wherein T is the number of seed crystals added per the weight of tris-(2,3-epoxypropyl)-isocyanurate in the reaction solution (number/g), m is the weight (g) of seed

Q1
cont
crystals added, D is the average particle size of seed crystals which is from 2 to 300 μm , and M is the weight (g) of tris-(2,3-epoxypropyl)-isocyanurate in the reaction solution.

5 12. The process according to Claim 8, wherein the seed crystals added in step (C') is β -form tris-(2,3-epoxypropyl)-isocyanurate, or a mixture of β -form tris-(2,3-epoxypropyl)-isocyanurate and α -form tris-(2,3-epoxypropyl)-isocyanurate.

10 13. The process according to Claim 8, wherein ultrasonic waves are applied to the liquid in the process of gradually cooling the liquid in step (C').

14. The process according to Claim 8, wherein the washing in step (D) is carried out by using a solvent
15 capable of providing a solubility of at least 0.5 g/100 g at 20°C to α -form tris-(2,3-epoxypropyl)-isocyanurate and a solubility of less than 0.5 g/100 g at 20°C to β -form tris-(2,3-epoxypropyl)-isocyanurate, in an amount of from 0.5 to 10 times by weight relative to the β -form
20 tris-(2,3-epoxypropyl)-isocyanurate crystals.

15. The process according to Claim 8, wherein the average particle size of the crystals obtained in step (C') is from 20 to 500 μm , and the drying in step (D) is carried out under atmospheric pressure or under reduced
25 pressure in a gas stream at a temperature of from 120 to 140°C.

16. The process according to Claim 8, wherein the

a'
cont

average particle size of the crystals obtained in step (C') is from 10 to 20 μm , and the drying in step (D) is carried out under atmospheric pressure or under reduced pressure in a gas stream at a temperature of from 40 to 120°C.

17. A process for producing β -form tris-(2,3-epoxypropyl)-isocyanurate crystals containing from 2 to 15 wt% of α -form tris-(2,3-epoxypropyl)-isocyanurate in the interior of the crystals, which comprises the following steps (A), (B), (C'') and (D):

(A) a step of reacting cyanuric acid with epichlorohydrin to form an addition product of cyanuric acid and epichlorohydrin, followed by dehydrochlorination to obtain a reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate,

(B) a step of removing epichlorohydrin from the reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate obtained in step (A), and dissolving the obtained tris-(2,3-epoxypropyl)-isocyanurate in a solvent,

(C'') a step of heating the liquid obtained in step (B) to a temperature of at least the temperature at which the liquid forms a saturated solution, thereafter cooling the liquid to a temperature lower by from 5 to 20°C than the temperature at which the liquid forms a saturated solution, and adding seed crystals thereto, and then gradually cooling the liquid at a cooling rate within 20°C/hr for crystallization, followed by filtration to

obtain crystals, and

(D) a step of washing and drying the crystals obtained in step (C").

18. The process according to Claim 17, wherein step (A) is a step of reacting (a) 1 mol of cyanuric acid, (b) from 5 to 180 mols of epichlorohydrin and (c) from 0.001 to 0.1 mol of at least one compound selected from the group consisting of a tertiary amine, a quaternary ammonium salt, a quaternary ammonium base, a tri-substituted phosphine and a quaternary phosphonium salt, as a catalyst, to obtain a reaction solution, adding from 2 to 6 mols of an alkali metal hydroxide or an alkali metal alcoholate to the reaction solution for dehydrochlorination, and then removing the resulting alkali metal salt to obtain a reaction solution containing tris-(2,3-epoxypropyl)-isocyanurate.

19. The process according to Claim 17, wherein the solvent in which tris-(2,3-epoxypropyl)-isocyanurate is dissolved in step (B) is acetonitrile, toluene, dioxane or dimethylformamide.

20. The process according to Claim 17, wherein the addition of seed crystals in step (C") satisfies the following formulae (1) and (2):

$$1 \times 10^{10} \geq T \geq 1 \times 10^2 \quad (1)$$

$$T = 1.4 \times 10^{12} (m/(M \times D^3)) \quad (2)$$

wherein T is the number of seed crystals added per the weight of tris-(2,3-epoxypropyl)-isocyanurate in the

Q1
Cont

reaction solution (number/g), m is the weight (g) of seed crystals added, D is the average particle size of seed crystals which is from 2 to 300 μm , and M is the weight (g) of tris-(2,3-epoxypropyl)-isocyanurate in the

5 reaction solution.

21. The process according to Claim 17, wherein the seed crystals added in step (C") is β -form tris-(2,3-epoxypropyl)-isocyanurate, or a mixture of β -form tris-(2,3-epoxypropyl)-isocyanurate and α -form tris-(2,3-epoxypropyl)-isocyanurate.

10

22. The process according to Claim 17, wherein ultrasonic waves are applied to the liquid in the process of gradually cooling the liquid in step (C").

23. The process according to Claim 17, wherein the washing in step (D) is carried out by using a solvent capable of providing a solubility of at least 0.5 g/100 g at 20°C to α -form tris-(2,3-epoxypropyl)-isocyanurate and a solubility of less than 0.5 g/100 g at 20°C to β -form tris-(2,3-epoxypropyl)-isocyanurate, in an amount of

15

20 from 0.5 to 10 times by weight relative to the β -form tris-(2,3-epoxypropyl)-isocyanurate crystals.

24. The process according to Claim 17, wherein the average particle size of the crystals obtained in step (C") is from 20 to 500 μm , and the drying in step (D) is

25 carried out under atmospheric pressure or under reduced pressure in a gas stream at a temperature of from 120 to 140°C.

25. The process according to Claim 17, wherein the
average particle size of the crystals obtained in step
(C") is from 10 to 20 μm , and the drying in step (D) is
carried out under atmospheric pressure or under reduced
5 pressure in a gas stream at a temperature of from 40 to
120°C.

A'
conf

add
2d

25. The process according to Claim 17, wherein the
average particle size of the crystals obtained in step
(C") is from 10 to 20 μm , and the drying in step (D) is
carried out under atmospheric pressure or under reduced
5 pressure in a gas stream at a temperature of from 40 to
120°C.